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Transmutation Engineering International Contributions Report

Introduction

Transmutation engineering provides the critical research and development necessary to support the Advanced Fuel Cycle Initiative (AFCI) and Generation IV reactor technologies. The top-level objective in this technical area is to provide development and engineering support for the transmutation of minor actinides and long-lived fission products so that informed decisions can be made in the next five years on the specific technologies and a path forward can be developed for implementation. In support of that objective, proof-of-principle information needs to be developed. In the near term five-year period, transmutation engineering activities are focused in the areas of physics, materials, and accelerator-driven systems. Subsequent to the decision on transmutation technologies and a successful proof-of-principle phase, engineering development and demonstration will be performed to provide proof-of-performance in preparation for deployment of transmuter technology.

Transmutation physics activities include development of computer programs (or codes), experimental measurements of cross-sections and reactivity feedback coefficients, evaluation of cross-sections for inclusion in nuclear data files, and performance of benchmarks and validations. Transmutation materials activities are divided into two parts: the development, testing, and modeling of structural materials to be used in the transmuter (either a Generation IV fast reactor or possibly an ADS), and research and testing of coolant technologies that can be used in these options. Transmutation ADS activities include the development and testing of target technologies, physics and engineering of coupled (accelerator/multiplier) systems, development of the safety case, and development of operation strategies. This research is being accomplished primarily



through participation in international programs and projects where significantly more ADS-related research is being performed.

To accomplish transmutation engineering tasks in an efficient manner, we have entered into several international collaborations. Many countries have extensive programs in fuels, materials, neutron source development, coolant technology, and coupled system engineering in which we also have an interest. It only makes good business sense that we should enter into collaborations to complement each other's work and make our collective research dollars stretch as far as possible. This report includes a summary of our international collaborations. The report covers activities in ADS coupling, ADS target research, physics code development, physics integral data experiments, structural materials, coolant technology, and accelerator research. An additional section summarizes the collaborations that University researchers (under contract to the national laboratories and receiving direct funding from the DOE) have entered into. Each section below provides a general description of the activity, the US contribution to the collaboration, the benefit of the collaboration, and an estimate of the cost of that benefit.

1 ADS COUPLING EXPERIMENTS

1.1 MUSE

1.1.1 Overall Description

The MUSE (**M**ultiplication avec **S**ource **E**xterne) program is a series of experiments to study the neutronics of ADS (accelerator-driven systems). These experiments have been carried out at the CEA-Cadarache MASURCA facility since 1995 in the 5th European framework plan. It began with the coupling of a Cf source in MASURCA, and was followed by a commercial (d,T) source. In 2001, a specially constructed (d,D)/(d,T) neutron generator (GENEPI) was placed in MASURCA, and the MUSE-4 program commenced. Different configurations and subcritical levels are being studied using the

MASURCA reactor. The core, in the central zone, is based on a uranium-plutonium MOX fuel. It includes in some configurations an inner region of lead and a tritium or deuterium target. Surrounding the core is a reflector region composed of a mixture of sodium and steel, and after this a shielding region built exclusively of steel.

The MUSE (phase) 4 experiments do not use a spallation source. Instead, an high-intensity pulsed-neutron generator GENEPI, constructed by CNRS/ISN/Grenoble, is being used to accelerate a 250-KeV deuteron beam towards either a deuteron target (d,d) or a tritium target (d,t) to produce well characterized neutron sources via fusion reactions: (d,d)-reactions produce neutrons with energies between 2 and 3 MeV, while the (d,t)-reactions produce neutrons between 13 and 15 MeV.

Measurements and validation of experimental techniques include:

- Subcriticality measurements by MSM (Modified Source Multiplication),
- Validation of time-dependent reactivity measurements (Pulsed Neutron Source, Feynman, and Rossi α),
- Source importance,
- $\beta_{\text{eff}}/\Lambda$,
- Spectrum (passive with foils, and active with He-3 detector) and spectral indices, and
- Reaction rate profiles.

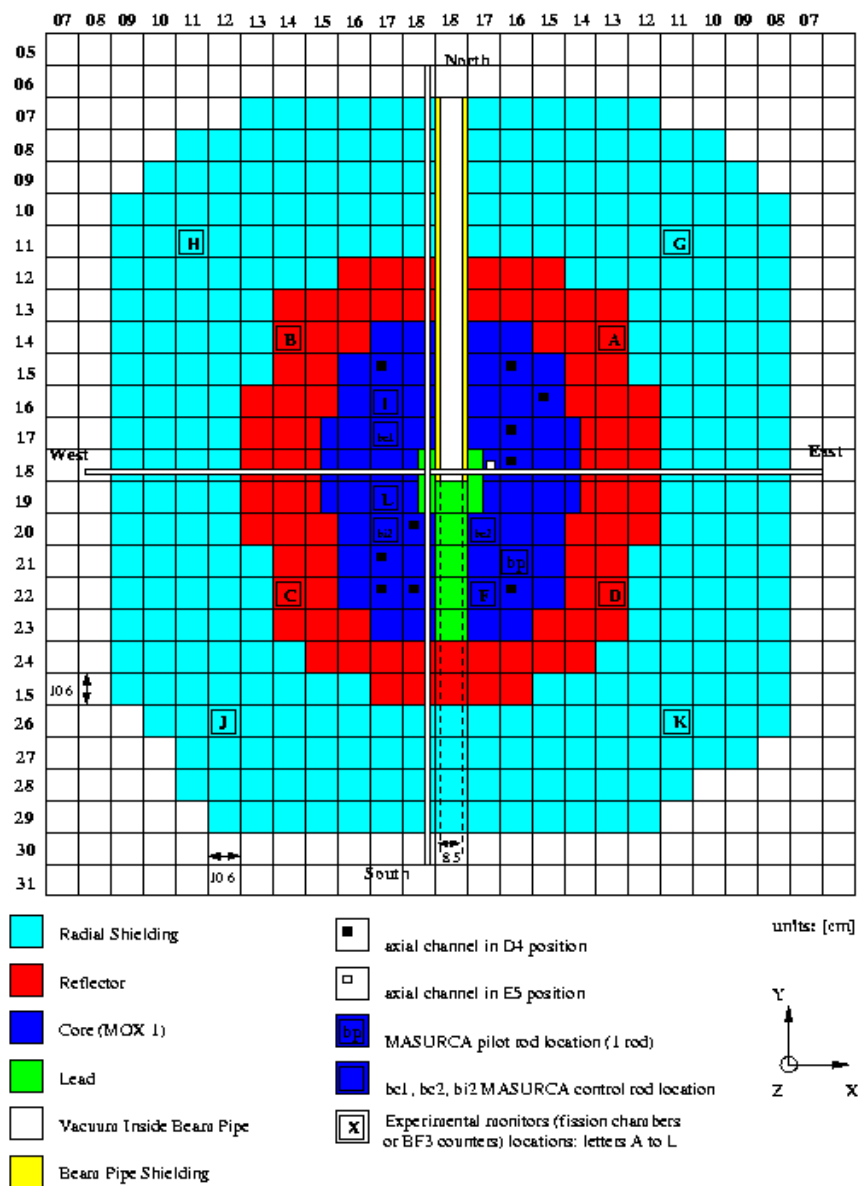


Figure 1. MUSE4 SC0 configuration.



1.1.2 Our Contribution

The US contribution to the MUSE experimental program include:

Experimental Activity. G. Imel (ANL) is head of the experimental group at the MASURCA reactor. He provides leads for the MUSE neutronic measurements and devises and validates the new experimental techniques.

Analysis Activity. G. Aliberti and G. Palmiotti (ANL) perform the analysis of the experimental results using different methodologies and basic data files. This activity provides the validation for methods and data in use in the USA for the neutronic calculations of accelerator-driven systems.

1.1.3 Benefit to AFCI

The MUSE experimental program validates the physics of accelerator-driven systems. The ADS concept for transmutation of nuclear waste is one of the options for the Series II transmutation systems that are to be selected by the Advanced Fuel Cycle Initiative (AFCI) program in 2007.

1.1.4 Estimate of Cost Benefit

The MUSE program has a cost over the year of several tens of millions of dollars. Participating to the MUSE program with a very limited cost (up to now less than a million dollar) has allowed the US to have access to invaluable data needed for the physics validation of ADS. It is also worth mentioning that in the Advanced Accelerator Applications (AAA) program of last fiscal year, the possibility to perform at the ZPPR facility (ANL-West), an experimental program with measurements similar to MUSE, was evaluated. The cost for that program was estimated on the order of \$30 million.



1.2 TRADE

1.2.1 Overall Description

The **TRADE** (**TRIGA Accelerator-Driven Experiment**) experiment, to be performed in the TRIGA reactor of the Italian ENEA-Casaccia Centre, consists in the coupling of an external proton accelerator to a target to be installed in the central channel of the reactor scrambled to sub-criticality. Aimed at a global demonstration of the ADS concept, this pilot experiment is based on an original idea by Carlo Rubbia.

The TRADE experiment is intended to validate the coupling of the different components of an ADS in a significant environment in terms of power of the global installation, using an existing critical reactor to be adapted to the objectives. In this way, the experiments of relevance that can be carried are:

- The dynamic behavior of an ADS, and in particular the possibility to operate at some hundred kW of power and at different subcriticality levels ($0.90 \leftrightarrow 0.99$),
- Subcriticality measurements at significant power,
- Correlation between reactor power and proton current,
- Reactivity control by different means and possibly by neutron source importance variation, and
- Compensation of power effect of the reactivity swing with control rods movements or with proton current variation.

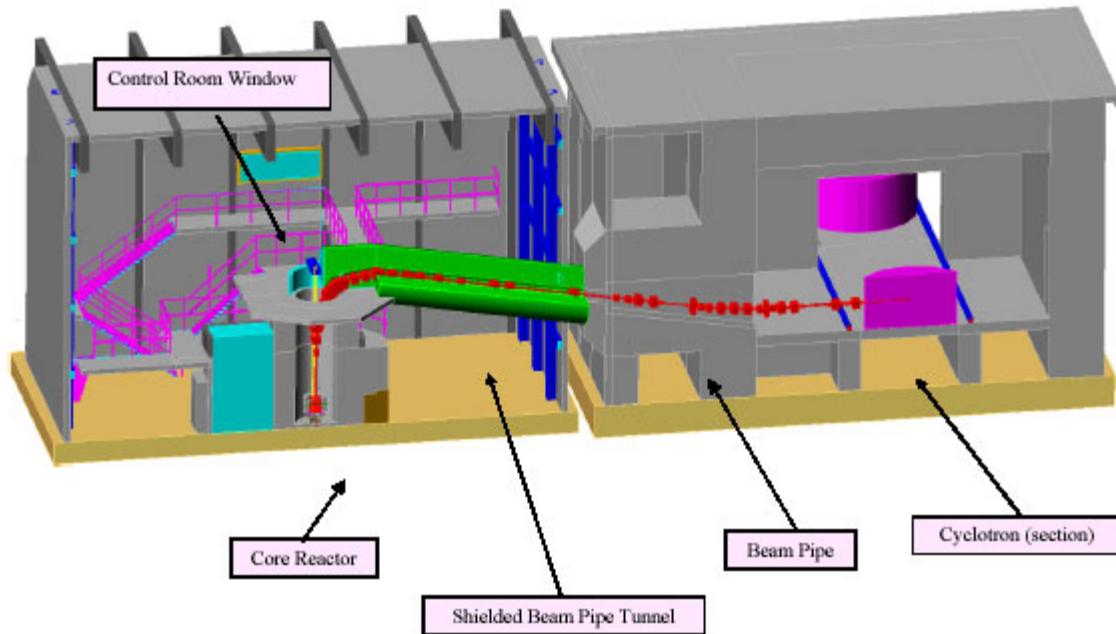


Figure 2. TRADE layout.

1.2.2 Our Contribution

The US contribution to the TRADE experiment is in three main fields:

Coordination. Max Salvatores (ANL) coordinates several areas of the project and organizes monthly meetings among to the international partners of the experiment.

Experimental Activity. G. Imel (ANL) is the lead experimentalist for TRADE. He proposes, designs, and performs the neutronic measurements that are carried out at the TRIGA reactor.

Analysis Activity. D. Naberejnev and G. Palmiotti (ANL) provide analytical support for the justification and design of the experiment.



1.2.3 Benefit to AFCI

The TRADE experiment will demonstrate the feasibility and operability of the ADS concept for the transmutation of nuclear waste. The ADS is one of the options for the Series II transmutation systems that are to be selected by the AFCI program in 2007.

1.2.4 Estimate of Cost Benefit

The TRADE experimental program cost has been evaluated on the order of \$60 million. The DOE contribution to the program is estimated to be about \$3 million, so the significant return on the investment is evident. It is also worth mentioning that in the AAA program of last fiscal year, the possibility to perform at the TREAT facility (ANL-West), an experimental program with goals similar to TRADE, was evaluated. The cost for that program was estimated on the order of \$15 million.

1.3 TRADE Target

1.3.1 Overall Description

The TRIGA Accelerator Demonstration Experiment (TRADE) couples a 100-MeV accelerator with a TRIGA reactor. A water-cooled heavy-metal target is placed at the end of the accelerator, in the center of the reactor to produce neutrons.

1.3.2 Our Contribution

Stuart Maloy and Eric Pitcher (LANL) are involved in the material choice for the heavy metal target. This involved holding a workshop at LANL in November 2002 and attending a workshop in May 2003 in Karlsruhe, Germany. They were also involved with constructing a matrix to compare four target options, tungsten, Inconel 718-clad tungsten, tantalum-clad tungsten, and tantalum. From this matrix, tantalum was chosen as the option for the target material.



1.3.3 Benefit to AFCI

The benefit to AFCI is in the accelerator-driven transmuter option; a solid target is a possible option. In addition, for the primary option (liquid LBE), tantalum is a candidate container material. Gaining the experience through collaboration with this target design will be useful information for designing a target for the ADS transmuter.

1.3.4 Estimate of Cost Benefit

2 ADS TARGET

2.1 MEGAPIE

2.1.1 Overview

MEGAPIE is a high-power (1 MW nominal) liquid-metal (lead-bismuth eutectic, or LBE) spallation neutron target under development at Paul Scherrer Institute (PSI) and supported by partners in Germany, France, Italy, Korea, and Japan, as well as the DOE. Liquid-metal targets are of particular interest to all partners, primarily because of the potential to operate at much higher incident proton flux than solid targets while offering significantly lower parasitic absorption of neutrons (higher neutron production ratios). Any future use of spallation targets by the AFCI project will benefit from the development of this technology.

MEGAPIE will operate in the PSI SINQ cyclotron facility. SINQ is a 580-MeV, 1.25-mA proton source, with near-term plans to increase current capacity. Commissioned about 10 years ago, this facility was designed with the intent of utilizing liquid metal targets; but until now there has not been sufficient technology development. The desire to operate at higher currents, coupled with the broad international interest in liquid metal targets, led to the concept of the MEGAPIE project. Once operational, MEGAPIE will be the first high-power target of this type.



The design phase of MEGAPIE is currently ending and the target fabrication is beginning. Ancillary systems are also in the fabrication phase. All ancillary systems and the target are scheduled for delivery to PSI by May 2004. A one-year integral test and commissioning phase will be then be performed. A three- to nine-month irradiation test is scheduled to start in the summer of 2005, with the duration to be determined by the responsible facility personnel based on confidence in materials and technology.

2.1.2 Our Contribution

The primary DOE contribution to MEGAPIE is \$800,000 over four years and one FTE engineering support on site at PSI. Additional DOE laboratory personnel have contributed in the areas of neutronics analysis, materials technology, and LBE technology. The engineering contribution includes analysis of components and accident scenarios, experimental work on the leak detector and heaters, support for preparations for integral tests, systems analysis using TRAC, and a comprehensive reliability study. A brief summary of the technical contributions of most of the participants is given in the following table.

Table 1. Collaborator Technical Responsibilities and Contributions (partial list)

Participant	Contribution
PSI	Project management, project integration, design support and approval, integral testing and in-beam testing
DOE	Engineering support (analytical and experimental), materials expertise and data (including PIE), neutronics analysis, reliability analysis.
CEA	Neutronics analysis, materials testing, heat exchanger structural analysis, neutron detectors.
FZK	Window cooling tests, LBE technology, materials technology, EMP analysis.
ENEA	Fill and drain system design and fabrication, heat exchanger tests, LBE freezing experiments, materials technology
Subatech	Overall design of target
Ansaldo	Cover gas system and heat removal system design and fabrication
JAERI	Materials and LBE technology
IPUL	Design, test and fabrication of EM pumps and flow meters.



2.1.3 Benefit to AFCI

The use of spallation targets, coupled to an accelerator, to produce neutrons to drive nuclear reactions for waste reduction and power generation is a viable part of the AFCI program. The production and testing of this next-generation target is a major technology development effort. By teaming with the MEGAPIE partnership, DOE benefits from the following primary technology developments and activities:

- 1 LBE-cooled target window development and testing in a high proton flux environment, including both materials and cooling technology development.
- 2 Development and demonstration of essential LBE loop components: pumps, flow meters, heat exchangers, preheaters, instrumentation, etc.
- 3 Development of a heat removal system capable of removing 600-kW waste heat and operating through as many as 10,000 beam trips to zero power instantaneously, and the development and testing of suitable control systems.
- 4 The demonstration of an organic coolant in a spallation target heat removal system, operating at high temperature but low pressure.
- 5 Measurement of gas production and development of methods for safe handling and release of gaseous byproducts from a liquid-metal target.
- 6 Demonstration of and data from neutron detectors placed near the heart of a high-power production target.
- 7 Validation of neutronics codes used to predict neutron yield, materials damage, and spallation products.
- 8 Development of techniques for post-irradiation handling and investigation of liquid-metal targets.
- 9 Post irradiation materials and component performance data.

Additional benefits to AFCI include lessons learned getting to final design and testing, including the extensive design work, preliminary component testing and optimization, and integral tests.

2.1.4 Estimate of Cost Benefit

The direct cost of MEGAPIE is €7.5 million (\$8.3 million). This is in direct payments to contractors for design and fabrication of parts. The cost of personnel at PSI and all



partner facilities, facility operational costs, and post-irradiation examinations are not included. There are more than 100 persons involved in MEGAPIE, with approximately 60 persons doing the majority of the work. Estimating 30 FTE per year, the manpower cost would be about \$9 million per year at a DOE laboratory. Project duration is five to six years, prior to post-irradiation examinations.

Facility costs for an equivalent experimental target test at Los Alamos National Laboratory (LANL) can be based on cost estimates conducted for the Russian-made target intended for LANSCE Area A. Including the necessary facility upgrade costs, facility personnel costs, and electricity, the estimate for a six-month irradiation was \$20 million to \$30 million.

Total cost of a MEGAPIE equivalent experiment at a DOE facility would then be about \$90 million, plus post-irradiation examinations.

2.2 MEGAPIE Post Irradiation Examination

2.2.1 Overall Description

The MEGAPIE project will be the first large-scale LBE target used for neutron production. The materials used for containing this LBE target will experience prototypic AFCI ADS transmuter conditions of LBE corrosion, stress, and proton and spallation neutron irradiation. Thus, a careful post-irradiation examination, Post-Irradiation Examination, of this target will provide valuable one-of-a-kind data to the AFCI project.

2.2.2 Our Contribution

Involvement of Stuart Maloy (LANL) in the MEGAPIE Post-Irradiation Examination is in a working group that will consist of attending meetings in Europe (possibly annually) and contribution to documents relating to the plans for careful examination of the target. Eventually, this will also involve transporting materials to LANL to be tested in the LANL CMR hot cells.



2.2.3 Benefit to AFCI

The MEGAPIE Post-Irradiation Examination will only provide valuable data if the target is carefully cut apart after irradiation without destroying the microstructure formed during irradiation. Thus, our involvement in the working group will help assure that quality data are obtained. These one-of-a-kind data are related to prototypic conditions in an ADS transmuter, which is needed information for the 2007 decision.

3 PHYSICS CODE DEVELOPMENT

3.1.1 Overall Description

The MCNPX code is the international standard radiation transport code for spallation source simulation. It is used in more than 100 institutions worldwide. Above the energy region in which tabulated evaluated nuclear data are available (150 MeV or below), physics models are used to describe nuclear reactions, with products of the reactions derived in a stochastic manner. To date, three intranuclear cascade models have been available in MCNPX to simulate high-energy hadron-nucleon interactions: the traditional Bertini model (the default model in MCNPX), the ISABEL model of Yariv, and the more recent CEM model of Mashnik.

3.1.2 Our Contribution

Through the DOE-CEA collaboration within the AFCI program, work was completed this fiscal year to add a new intranuclear cascade model to MCNPX—the INCL model of Cugnon. In addition, the evaporation model of Schmidt, ABLA, which has been developed in concert with the INCL model, has been added. This effort required close collaboration between researchers at CEA-Saclay, who provided the computer routines, and scientists at Los Alamos National Laboratory, who incorporated the routines into the MCNPX code. Finally, simple test problems were run to demonstrate that the INCL/ABLA models, as implemented in MCNPX, worked as expected.



3.1.3 Benefit to AFCI

The strongest feature of the combined INCL/ABLA model is its ability to predict nuclide yields produced by spallation. Comparison of the predicted yields from the four different intranuclear cascade models to experimental data shows the new INCL/ABLA model gives the best general agreement. Incorporation into MCNPX makes the model available to a large user base who can test the model in a broad spectrum of applications and compare its results to other intranuclear cascade models.

3.1.4 Estimate of Cost Benefit

The development and validation of the new INCL and ABLA physics models is an ongoing effort involving primarily three institutions with approximately six full-time equivalent employees dedicated to these activities. The models have been in development for approximately five years, for a total of approximately 30 person-years invested in these new physics models. To carry out a parallel effort at a DOE laboratory would cost approximately \$7 million. Through the DOE-CEA collaboration, these physics models are being made available to the DOE at no cost.

4 INTEGRAL DATA EXPERIMENTS

4.1 PROFIL and TRAPU

4.1.1 Overall Description

The PROFIL and TRAPU programs were irradiations experiments performed at the French fast reactor PHENIX in the 1970s and 1980s and were intended for gathering information on cross sections and transmutation rates of actinides for fast-spectrum systems.

During the PROFIL irradiation campaign, one or two standard pins, with separated capsules (46 in PROFIL-1, 2x42 in PROFIL-2) containing uranium, plutonium,



neptunium, americium, and curium isotopes, were located in a standard subassembly in the first rows of the inner core of the PHENIX reactor, far away from neutronics perturbations, in order to obtain clean irradiation conditions (e.g., stability of the neutron flux in the region close to the center of the reactor). Following the in-reactor irradiation, mass spectroscopy was then used, with simple or double isotopic dilution and well-characterized tracers to measure concentrations.

The TRAPU experiment consisted of a six-cycle irradiation of mixed-oxide pins that contained plutonium of different isotopic compositions but heavily charged in the higher isotopes (Pu-240, Pu-241, and Pu-242). Standard pins were placed in standard PHENIX subassemblies and irradiated in positions close to the center of the reactor. Three types of plutonium-containing pins were used. After irradiation, small samples (20 mm high) were cut from the experimental pins (both fuel and clad) and put into a solution to determine the fuel composition by nuclide. Neodymium-148 was used as the burn-up indicator because it is a stable fission product with a small capture cross section, and it enables determination of the number of fissions that have taken place in the sample.

Experimental results on PROFIL and TRAPU were obtained in the framework of the AFCI DOE/CEA agreement in exchange of the ZPR-9 ANL experimental data for the simulation of fast gas-cooled reactors.

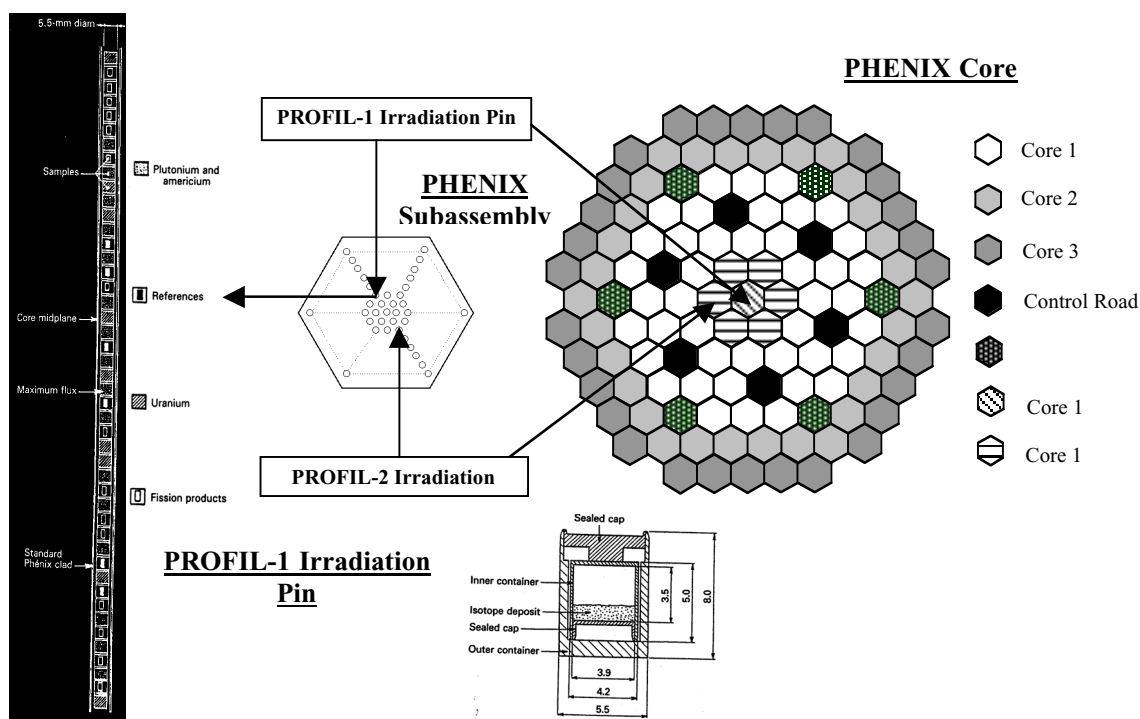


Figure 3. Stainless steel double container for PROFIL irradiation.

4.1.2 Our Contribution

G. Palmiotti and M. Salvatores (ANL) perform the analysis of the irradiation experimental results of PROFIL and TRAPU. The information that can be gathered from the PROFIL post-irradiation analysis is related to the evaluation of the reaction rates [mainly capture and (n,2n) rates] for a given isotope. In particular, the analysis of the experiment is based on the relation existing between the burn-up dependent variations of the atom number densities and the microscopic cross sections. The TRAPU experiments provide information not only on capture cross sections but also on some fission cross sections and half-life decay constants.

4.1.3 Benefit to AFCI

The experimental data of the PROFIL and TRAPU irradiation experiments provide very clean and useful information on both cross section data and transmutation rates of actinides. These data are essential for the validation of the methods and data to be used in



the Series II phase of the AFCI program in which transmuter systems will be used to reduce the existing inventory of nuclear waste.

4.1.4 Estimate of Cost Benefit

The cost of the PROFIL and TRAPU irradiation experiments can be evaluated in several millions of dollars. The past and future budgets of DOE spent for the analysis of the integral experiments for cross section validation is well under \$1 million. For such a small amount plus the exchange of the ZPR-9 data, DOE has access to data that, in view of their valuable usefulness for the characterization of transmutation rates of higher plutonium isotopes and minor actinides, have been considered proprietary by CEA.

4.2 MALIBU

4.2.1 Overall Description

The MALIBU (**MOX And LEU Fuels Irradiated to High Burn-up**) is an international program under the direction of Belgonucleaire to perform detailed isotopic measurements of high-burn-up UOX and MOX fuels from pressurized and boiling water reactors. The goal of the program is to provide high-quality isotopic data on UOX and MOX fuels that can be used for several applications, including code and data validation, burn-up credit, transmutation, high-burn-up fuel development, BWR void effect, source terms, and radioprotection. The interest on the part of the AFCI program is for the validation of codes and data for applications involving deep-burning of fuel, which would occur in a transmutation-recycle program. The work of the experimental program will be performed over several years; a schedule is provided in the table below.

Table 2. MALIBU Program Schedule as of June 2003

Feasibility study, Program preparation	2001 - 2002
PWR Fuel (Goesgen)	
Extraction and transfer to laboratory	2002
Characterization and irradiation reporting	2003
BWR Fuel (Gundremmingen)	
Extraction and transfer to laboratory	2002
Characterization and irradiation reporting	2003
Program Startup	March 2003
First PCM and official program opening	June 2003
Sample cutting, conditioning, and transfer between laboratories	2003
Radiochemical measurements (SCK.CEN, PSI, ITU, CEA)	2003 - 2004
Analysis and reporting	2004 - 2005

As previously stated, MALIBU is an international program involving several organizations. As of the first Program Committee Meeting in June 2003, there are eight participating organizations: The Paul Scherer Institute (Switzerland), Electricity de France (France), CEA/Cogema (France), Oak Ridge National Laboratory (US), RWE (Germany), Kerndraftwerk Göesgen (KKG, Germany), SCK.CEN (Belgium), and Belgonucleaire (Belgium). The participants provide both financial and work-in-kind support to the program in exchange for sharing the data that are produced. The participating organizations form a Program Committee, which is a voting body that provides direction for the experimental program.

Of particular importance is the selection of the fuel samples that will be used for the experimental program. The fuel samples that have been obtained include LEU and MOX fuel rods from 9 x 9 BWR LEU and MOX bundles and 15 x 15 LEU and MOX PWR

assemblies. The fuel rods were chosen such that the fuel burn-ups are very high, generally about 70-80 GWd/tM, which generally exceeds current fuel peak burn-ups of about 60 GWd/tM. The BWR assemblies were irradiated in the Gundremmingen reactor and the PWR assemblies were irradiated in the Göesgen reactor. The characteristics of the fuel assemblies are given in the table below. These fuel samples will be used for the base program that will perform the full characterization of the fuel consisting of the measurement of 17 actinides and 34 fission products. An extended program, not currently included in the program, has been proposed for the reduced characterization of the UO_2 BWR fuel at different axial levels to example the impact of the coolant void on the isotopic content. This reduced characterization covers 15 actinides and 20 fission products that have been shown to be sensitive to the void level. The following figure gives the list of isotopes selected for measurement.

Table 3. MALIBU Program Fuel Sample Specifications

	Göesgen PWR			Gundremmingen BWR		
Sample	GGM1	CGU1	CGU2	GRM1	GRM2	GRM3
Fuel Type	MOX	UO_2	UO_2	MOX	MOX	MOX
Pu_{fis} (wt. %)	5.5	—	—	5.5	5.5	5.5
U235 (wt. %)	depleted	4.3	4.3	depleted	depleted	depleted
End of Irradiation	July 2001	July 2001	July 2001	July 2001	July 2001	July 2001
Sample Burn-up (GWd/tM)	~70	~70	~50	~80	~75	~50
Sample Axial Position	Peak BU	Peak BU	Top	Peak BU	Mid	Top

The radiochemical measurements will be performed at four laboratories: PSI, ITU, SCK.CEN, and CEA. The measurements will be performed using well-established mass spectrometry methods, including ICP-MS and TIMS. The schedule of measurements calls for each sample being measured independently by two laboratories, and one sample being



measured by three laboratories. Previous experience with the ARIANE program has shown that these cross-checks provide confidence in the results and an indication of the uncertainty levels.

4.2.2 Our Contribution

As a participant of the MALIBU program, the contribution of the AFCI program includes financial support, program review and direction, and computational analysis to compare with measurements. A program such as MALIBU requires substantial financial investment to pay for the program expenses. The AFCI program direct financial support to the MALIBU program is €334,000 as of November 2001, or approximately \$390,000. As a participating member, ORNL provides a review of the program to ensure that the objectives will be met and that the data meets the requirements of the AFCI program. This review and direction is provided by ORNL as a voting member of the MALIBU Program Committee and at annual review meetings at which the progress of the program is discussed. Finally, computational analyses of the fuel irradiation will be performed by ORNL to compare with the experimental results. These calculational analyses will not only be the basis of the methods and data validation, but will also be presented to the other participating members of the program for comparison with their analyses.



<u>BASE ACTINIDES</u>	
^{234}U , ^{235}U , ^{236}U , ^{238}U	^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu
<u>MINOR ACTINIDES</u>	
^{237}Np	^{241}Am , $^{242\text{m}}\text{Am}$, ^{243}Am ^{242}Cm , ^{243}Cm , ^{244}Cm , ^{245}Cm
<u>FISSION PRODUCTS</u>	
Volatiles:	^{129}I , ^{133}Cs , ^{134}Cs , ^{135}Cs , ^{137}Cs
Metallics:	^{90}Sr , ^{95}Mo , ^{99}Tc , ^{101}Ru , ^{106}Ru , ^{103}Rh , ^{109}Ag , ^{125}Sb
Lanthanides:	^{144}Ce , ^{147}Pm , ^{155}Gd ^{142}Nd , ^{143}Nd , ^{144}Nd , ^{145}Nd , ^{146}Nd , ^{148}Nd , ^{150}Nd ^{147}Sm , ^{148}Sm , ^{149}Sm , ^{150}Sm , ^{151}Sm , ^{152}Sm , ^{154}Sm ,

Figure 4. List of isotopes selected for the MALIBU program radiochemical analysis.

4.2.3 Benefit to AFCI

The primary benefit to the AFCI program is the access to a substantial amount of high-quality isotopic data for high-burn-up UOX and MOX fuels, at a relatively low cost. Using this data, the accuracy of methods and data will be established based on these experiments, which closely reflect the deep-burning that will occur in transmutation and recycling programs. The determination of the accuracy of the calculation of the isotopic inventories in the irradiated fuel will ensure that the material flows in the transmutation cycle are correct and that the accuracy of the prediction isotopic inventories are known. Hence, the major benefit to the AFCI program is ensuring that the inventory of spent UOX and of future recycled MOX fuel can be accurately predicted. An additional benefit to the program is the identification of additional methods and data needs required to carry out the program's mission.



4.2.4 *Estimate of Cost Benefit*

The total budget of the program as given at the June 2003 Program Committee Meeting is €2.385 million in November 2001 (approximately \$2.9 million in August 2003), which is split among the participating members. This cost does not include the cost of the radiochemical analyses to be performed by the four laboratories in lieu of financial contribution and, based on reductions in participation fees, represents an additional €700,000 (approximately \$800,000). Therefore, the total cost of all services is valued at approximately \$3.7 million. The cost to the AFCI program for participation in the program is €334,000 (approximately \$390,000). Additional support required for ORNL staff has been budgeted at \$220,000 for a total cost of \$610,000.

5 STRUCTURAL MATERIALS

5.1 STIP

5.1.1 *Overall Description*

The SINQ Target Irradiation Program (STIP) uses the 590-MeV SINQ proton accelerator to irradiate structural materials used for target containment. The program is coordinated by Yong Dai at the Paul Scherrer Institute. Specimens are placed in Zircalloy tubes, which replace some of the lead-filled target tubes used to produce neutrons. Each tube is approximately 10 mm in diameter and is instrumented with a thermocouple to record temperature during irradiation. Activation foil stacks are placed in select locations throughout the irradiation volume to determine the proton and neutron flux experience by the specimens during irradiation. There have been three STIP irradiations so far, and one more is planned to start in 2004. After this irradiation, the target will be replaced by MEGAPIE. The cost to the user is payment for specimens retrieval and shipment to his testing laboratory which is usually around \$30,000 to \$40,000.



5.1.2 Our Contribution

Our involvement in the STIP program has been three-fold:

- 1 Stuart Maloy (LANL) has contributed to STIP I by analyzing irradiated rods and STIP II, III and IV by providing specimens for irradiation (these specimens are not ready for analysis yet).
- 2 Mike James (LANL) and Wei Lu and Monroe Wechsler (NCSU) have also helped in determining the fluence of materials in STIP I by analyzing and performing calculations from the activation foil spectra and will also be helping with the analysis of foils from STIP II.
- 3 Stuart Maloy (LANL) has co-organized a workshop, International Workshop on Spallation Materials Technology (IWSMT), during which data from these STIP irradiations and other data related to spallation materials technology are discussed by researchers around the world. This workshop is held every 1.5 years, and the main organizing laboratories are LANL, Los Alamos, NM, USA; ORNL, Oak Ridge, TN, USA; PSI, Villigen, Switzerland; Forschungszentrum Julich, Julich, Germany; and KEK/JAERI, Japan. The workshop location alternates each year. The next workshop will be held in Hayama, Japan, in December 2003. The cost to each laboratory is about \$2,500 for each workshop.

5.1.3 Benefit to AFCI

Involvement in the STIP irradiations is a significant benefit to AFCI. These irradiations provide data on the effect of high-energy proton irradiation on the mechanical properties of AFCI structural materials at prototypical temperatures. There is only one other location in the world, LANSCE, where irradiation such as this could be performed, and presently the facility for performing these irradiations, the Los Alamos Spallation Radiation Effects Facility (LASREF), is not operational. Such data are crucial in determining materials design limits in an ADS transmuter relating to the 2007 decision.



5.1.4 Estimate of Cost Benefit

6 COOLANT TECHNOLOGY

6.1 LBE Handbook

6.1.1 Overall Description

The OECD/NEA chartered the LBE Expert Group, which includes members representing DOE/LANL (US), FZK (Germany), CEA/CNRS (France), ENEA/Ansaldo (Italy), PSI (Switzerland), SCK-CEN (Belgium), CIEMAT (Spain), KTH (Sweden), JAERI (Japan) and KAERI (S. Korea). The group had its first meeting in May 2003 at FZK; all member countries were present. During the meeting, we determined the scope of the LBE Handbook, which will include available data for lead. We determined the assignments, a draft schedule, and a list of recommended experts to review the draft. We also determined the scope and schedule for an international joint development plan for the LBE technology development.

6.1.2 Our Contribution

The draft US program plan, including a technology gap analysis and five-year major tasks and milestones, was submitted to the group as a template for the other member countries.

We have compiled the available Russian LBE materials data on corrosion and radiation effects into a report, and started compiling the international LBE corrosion test data to be supported by analysis and interpretation through corrosion modeling. These reports will be submitted to the LBE Handbook project.



6.1.3 Benefit to AFCI

In the AFCI Program, we continue to study and develop the LBE spallation target and transmuter coolant options. The LBE Expert Group members represent or have access to almost the entire LBE research community worldwide outside Russia. Having the expert input from these experts and the R&D organizations in the OECD member countries they represent significantly enhances the quality, completeness, and utility of the Handbook. Additionally the international joint development plan will perform to some degree technology-based integration and coordination, leveraging participating members' R&D investment, expertise, facilities, and human resources. The overall effect is an acceleration of the LBE technology maturation for better-informed technological selection decision.

6.1.4 Estimate of Cost Benefit

The AFCI supports US expert participation at the \$50,000/yr level. There are more than a dozen international experts in the group, each contributing their own expertise and that of the organizations and/or countries they represent. The simplest cost benefit estimate is ~\$600,000/yr by simple multiplication. The total number of LBE researchers we have better access to through this group numbers is more than 100 worldwide, compared to fewer than a dozen in the AFCI. There are also a number of significant facilities producing test data that are valuable to the AFCI Program.

7 ACCELERATOR

7.1 OECD/WPPT

7.1.1 Overall Description

As part of the Organization for European Community Development/Working Party on Partitioning and Transmutation (OECD/WPPT), we chair the Accelerator Utilization and



Reliability Subgroup (AURS). This group is made up of several leaders in the field of accelerator research.

The objective of the AURS is to produce a set of accelerator reliability guidelines that ADS designers can use as part of the conceptual design.

Specifically, the scope includes the following activities:

- Develop an expert opinion on what the fault and interrupt rates would be if an accelerator were to be designed and built with today's technology and with high-reliability as a primary objective. In order to do so:
 - Collect the fault and beam interrupt data from representative accelerators being utilized around the world for various applications (emphasis is on high-power machines in user facilities),
 - Analyze and sort to the extent possible the data by components or accelerator section resulting in failure, and by duration of the beam interrupt (groups of interest are less than a second, a few seconds, tens of seconds, and longer duration);

The main deliverable of the working group will be an accelerator reliability report.

Accelerator reliability is one of the central issues that must be resolved in order to eventually field a large-scale ADS machine. The accelerator must operate smoothly and reliably with as few beam trips as possible to reduce the transient stresses on the coupled subcritical multiplier. Accelerator faults that result in a beam interrupt on the order of a second or longer are not acceptable for accelerator-driven transmutation systems.

Frequent beam interrupts not only disrupt the transmutation and electricity production efficiency but result in thermal-fatigue in the target, multiplier, and cooling systems structure. Beam interrupts of different durations are important for different phenomena, but typically interrupts of very short duration (less than 100 ms) are inconsequential. Therefore, understanding the causes of the faults and improving the accelerator design to reduce the number of longer duration interrupts are of primary importance.



The accelerator also must be able to be shutdown on demand and, in many cases, automatically during emergency situations. To avoid power surges in the multiplier, the accelerator must provide physical constraints that eliminate overpower situations. Sensors that control the emergency shutdown must be *fail-safe*.

7.1.2 Our Contribution

Our contribution to the task activity was to summarize the accelerator technology that is currently available for use in an ADS. This information has been documented in a report that was generated in FY03. The working group meets twice per year. The cost of participating in the meetings and producing the report was approximately \$50,000.

7.1.3 Benefit to AFCI

The main benefit of this collaboration is in the information exchange among the participants, and in the eventual development of the safety case and operation plan for an ADS. Information exchange leads to a better understanding of the programs that are proceeding in Europe and Asia and provides a motivation for additional work.

7.1.4 Estimate of Cost Benefit

The cost benefit of this activity is approximately \$1 million per year. This estimate was derived by taking the number of experts working on this activity and estimating the full-time equivalents spent in performing the work.

8 UNIVERSITY PROGRAMS CONTRIBUTIONS TO INTERNATIONAL COOPERATION

8.1.1 Overall Description

Through multi-university and multi-laboratory collaborations, universities are conducting research at, in collaboration with, and/or for international organizations. Universities that have contributed include the following:



- University of Nevada Las Vegas (UNLV)
- Idaho State University (ISU)
- University of Michigan (Michigan)
- North Carolina State University (NCSU)

The international organizations involved in these collaborations with AFCI-supported university programs include:

- V.G. Khlopin Radium Institute, St. Petersburg, Russia (KRI)
- Karlsruhe Lead Laboratory, Germany (KALLA)
- Institute of Physics and Power Engineering, Obninsk, Russia (IPPE) and the International Science and Technology Center (ISTC)
- Imperial College of London, UK (ICL)
- Tbilisi University, Republic of Georgia (TU)
- Paul Scherrer Institute, Switzerland (PSI)
- Atomic Energy Commission of France, Cadarache, France (CEA)

8.2 Our Contribution

8.2.1 UNLV Transmutation Research Program

At UNLV, which has become the lead US university for transmutation research, two research projects involve interaction with international organizations. In addition, UNLV is building infrastructure in an international collaboration, and they have become a member of the ISTC. In one project, UNLV has contracted with KRI to develop a 60-element ^3He detection system surrounding a cylinder of lead. Neutron multiplicity measurement for target/blanket materials. This device was designed to be inserted into particle beams to measure the spatial and directional dependence of multiple-neutron generation events (neutron multiplicity) in accelerator target/blanket materials. This neutron multiplicity measurements system was constructed at KRI with \$112,500 of UNLV transmutation research program (TRP) funding, then it was shipped to UNLV, where it is being set up for calibration and validation. The estimated value of the



detection system is \$1 million. In addition, a UNLV student has visited KRI in St. Petersburg to collaborate with Russian scientists in modeling of systems for predicting measurement performance, and an \$11,100 follow-up contract has been written to support computational work.

In another UNLV TRP international collaboration, UNLV has assumed responsibility for an ISTC project, has taken over the contract from Los Alamos, and has acquired a lead-bismuth-cooled target for proton accelerators. The device, which has been named TC-1, is being installed in a laboratory at UNLV where it will be used for thermal hydraulic and material corrosion experiments. When the UNLV LBE lab has been completed, the device will be set up and qualified by scientists and engineers from IPPE in Obninsk, Russia. This work is supported with \$186,000 of UNLV TRP funds over 25 months. UNLV faculty members have also established a relationship with scientists at KALLA and have formed an international advisory committee on lead and lead-bismuth systems.

Finally, UNLV faculty and students are working with KRI to develop waste forms for the back-end of fluorine-based separations processes. One concept is the use of the mineral fluorapatite as a waste form following chemical processing of used nuclear fuel. Another concept is the immobilization of fission iodine by reaction with a Fullerene-containing carbon compound (FCC) and insoluble natural organic matrix (NOM). Researchers at KRI have proposed that the iodine-loaded FCC material, when combined with ceramics, is stable enough for use as a long-term storage form, and may also be a matrix target for transmutation of iodine. For this work UNLV has established a two-year project at KRI with \$152,000 per year.



8.2.2 Other University Programs

Idaho State University—Idaho Accelerator Center (ISU-IAC): The Idaho State University, which is the second largest component of the AFC university programs, is collaborating with students and scientists from two international organizations. In one project, students from Tbilisi University in the Republic of Georgia are working on a project to evaluate dose conversion coefficients for rare isotopes that are produced in high-energy spallation targets.

Another project has been initiated to use high-energy linear accelerators to drive nuclear assemblies in Dubna, Russia. This work involves accelerators and reactors at the KRI as well as more powerful accelerators that will be installed at ISU-IAC in the future. In addition, this project may involve measurements of pure isotopes at KRI.

8.2.3 LANL-Directed University Projects

Three universities have been involved in research to support LANL's requirements:

North Carolina State University: Faculty and students have been calculating radiation damage, including production of displacements, helium, hydrogen, and heavier transmutation products, as well as energy deposition in targets for generation of high-energy spallation neutrons. They are examining response in target materials, containment structures, and entrance windows of the target assemblies for the SINQ spallation neutron sources that are under design and development at the Paul Scherrer Institute. These targets include the SINQ Targets 3 and 5 as part of the SINQ Target Irradiation Program (STIP), which is an international collaboration that includes the France (CEA), Germany (FZJ), Japan (JAERI), Switzerland (PSI), and the US (LANL and ORNL). Target 3 was used for the STIP I irradiation, which ended in December 1999, and Target 5 is being used for the STIP III irradiation, which is scheduled to end in December 2003.

University of Michigan: Several faculty and students have compared neutronics tools to validate design methods; these codes have included those in use in international



programs. In addition, they have been studying dynamic behavior of accelerator-driven subcritical reactor systems, such as the recent MUSE experiments and the future TRADE program. This has involved development of dynamic models for simulating multiple pulses of spallation neutron sources and methods for determining the reactivity in ADS. Their emphasis has been on developing computational tools that can accurately and efficiently represent the localized nature of spallation sources in determining the power distribution and reactivity in transient conditions. A Michigan student developed numerical algorithms based on a two-dimensional time-dependent diffusion theory code that can accurately account for step changes in localized sources in time to establish a space-dependent dynamic model for simulation of ADS transient behavior. This involves separate treatments for the shape-function and amplitude-function calculations that can represent prompt space-time variations in neutron flux within the quasi-static formulation. These studies continued in the development of methods to account for the spatial dependence in reactivity measurements. The method was recently used to correct measurement results from the MUSE program.

Imperial College of London: One faculty member and one student are developing tools to model defects in nitride-based nuclear fuels under a \$60,000 contract for the AFCI Fuels Research program. In this international collaboration, a capability to model radiation-induced defects in surrogates will provide the capability to predict the performance of nitride fuels. In addition, the student from ICL worked at LANL during the summer of 2003.

8.3 Benefit to AFCI

8.4 Estimate of Cost Benefit

The estimated value of these university-international collaborations would be more than \$3 million for the same research, development, and construction of equipment to be conducted at US national laboratories.